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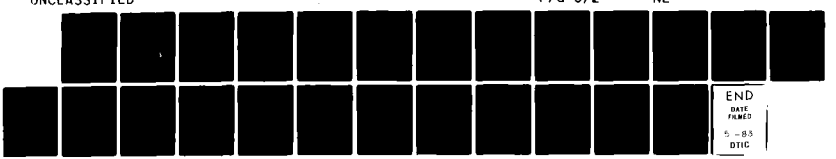

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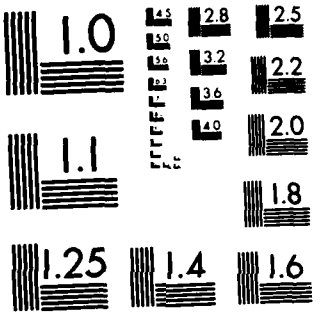

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This communication is the final report for ONR Research Contract N00014-81-C-0165, which extended over the period of January 1981 through November 1982. The general subject matter is development of high performance optical memories based upon photochemical hole burning.

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Contract N00014-81-C-0165
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FINAL REPORT

An engineering Study of Optical Memories Based on Photochemical Hole Burning

By

G. C Bjorklund

IBM Research Laboratory
San Jose, California 95193

March 22, 1983

AN ENGINEERING STUDY OF OPTICAL MEMORIES BASED ON PHOTOCHEMICAL HOLE BURNING

Final Report

Contents

I. INTRODUCTION

This communication is the final report for ONR Research Contract N00014-81-C-0165, which extended over the period January 1981 through November 1982. The general subject matter is development of high performance optical memories based upon photochemical hole burning.

II. RESEARCH ACCOMPLISHED

The initial phase of our engineering study of optical memories based upon photochemical hole burning (PHB) has yielded several significant accomplishments. The utility of FM spectroscopy for rapid and sensitive detection of photochemical holes was conclusively demonstrated, aggregate color centers in alkali halide crystals were identified as suitable PHB recording materials for exploratory work on memory applications, technically reasonable PHB memory systems configurations were devised for DASD-type applications, PHB materials sensitive to GaAlAs laser wavelengths were discovered, satisfactory GaAlAs laser tuning was achieved, and reading and writing of holes using GaAlAs diode lasers was explicitly demonstrated.

A. Detection of Photochemical Holes Using FM Spectroscopy

The basic principles of the FM spectroscopy technique were explained in detail in the original proposal. Reading of the data stored in the PHB memory is accomplished by probing the recording medium with frequency modulated (FM) laser radiation produced by passing the output of a single frequency laser oscillating at optical frequency ω_c through an external phase modulator driven at RF frequency ω_m. The transmitted light is incident on a fast photodetector connected to phase-sensitive electronics. A heterodyne amplified beat signal at ω_m is detected whenever the FM spectrum of the transmitted light is perturbed by the presence or absence of photochemical holes coincident with one of the FM sidebands.

Rapid detection of photochemical holes has been demonstrated using 200 MHz wide, 20% deep holes burned into the 6070Å zero phonon line of the N_1 color center in NaF (Section III, Publications, No. 1). Since the unbleached absorption of the peak of the zero phonon line was 10% of the laser light, the sidebands experienced a maximum differential absorption of about 1%. An actively stabilized ring dye laser provided tunable single

frequency radiation at ω_c with a (jitter) width of 1-2 MHz. The laser beam was frequency modulated with a $LiTaO_3$ phase modulator driven at ω_m=70 MHz. The beam emerging from the sample was detected with a P-I-N photodiode. The photodiode electrical output was amplified by a low noise amplifier, and the beat signal at ω_m homodyne detected by a double-balanced mixer.

The laser frequency ω_c was scanned over the photochemical hole with ω_m held constant and the strength of the best signal was monitored. Since ω_m was less than the hole width, the experimental lineshape observed was the derivative of the hole absorption profile. (This differentiation process serves to suppress the slowly varying background of the broad inhomogeneous line.) The experimental signal was easily detected in real time with 1 MHz bandwidth electronics, indicating that the hole could have been detected in 1 μsec. The 1 μsec detection capability was also explicitly demonstrated by chopping the laser beam into a string of widely separated μsec duration pulses and monitoring the FM beat signal produced during each pulse. When the laser frequency ω_c was tuned far off of the hole, a null signal was observed and when ω_c was tuned to the point of maximum slope of the hole absorption profile, easily detectable signals were obtained from each pulse.

The detection speed achieved in these experiments was limited to 1 μsec due to the relatively narrow bandwidth of the storage scope used to record the FM signals. If faster detection electronics had been utilized, a less than 100 nsec detection time could have been achieved for ω_m=70 MHz.

A major advantage of FM spectroscopy is that single mode lasers are essentially shot noise limited at RF and microwave frequencies. The ultimate achievable detection speeds are more than sufficient for memory applications. In fact, we have performed a rigorous signal

to noise analysis (Section III, Publications, No. 3) which shows that for $\omega_m \geq 1$ GHz, a 1% differential absorption should be detectable in less than 1 nsec under shot noise limited conditions.

B. PHB in Aggregate Color Centers Contained in Alkali Halide Hosts

The zero phonon lines of several types of aggregate color centers in alkali halide host crystals have previously been shown to demonstrate photochemical hole burning behavior. The alkali halide host materials are attractive for memory applications, since they are optically isotropic, are transparent to a wide range of wavelengths, polish reasonably well, can be obtained in large size samples with high purity and good optical quality, are stable at room temperature, and can withstand repeated thermal cycling to the 2K operating temperature.

We have surveyed the published literature on the spectroscopy of aggregate color centers in alkali halides and have identified several general trends (Section III, Publications, No. 2). In particular, we have found that the wavelengths λ of the zero phonon lines obey Mollwo-Ivey relations of the type

$$\lambda = K\, a^n$$

where K is a constant for each type of aggregate center, a is the lattice constant of the host, and n~1-2.

In principle, each type of aggregate color center can be found in each of the 20 alkali halide host crystals, which have a values, ranging from 2.0 to 3.9Å. Thus, if the assumption is made that the hole burning properties depend primarily on the type of aggregate color center and not on the particular host, it is possible to choose different materials for matching to the wavelength range of various sources with desired properties, *e.g.*, HeNe or GaAlAs.

In order to achieve the 10^{11} bits/cm^2 areal storage density possible in PHB memories, it is necessary to focus the laser beam to diffraction limited spot sizes on the order of 1 μm. The depth of focus associated with such small spot sizes is only on the order of a few μm and thus it is necessary to be able to produce the optically active species in thin films consistent with this depth of focus.

We have succeeded in producing such thin films of aggregate color centers by irradiation of bulk alkali halide crystal samples with 25 keV electrons or 100 to 200 keV ions (Section III, Publications, No. 2). The incident particles penetrate only a few μm, creating a high concentration of defect centers near the surface. Aggregation of the color centers occurs readily at room temperature. In many cases, the density of the aggregate centers in the thin film samples is sufficient for their zero phonon lines to be detected directly by absorption. The burning of 100 MHz wide photochemical holes was demonstrated in both electron and ion irradiated samples.

C. PHB Memory Systems Configurations

In addition to offering a potential gain of 10^3 in storage density, the phenomenon of photochemical hole burning allows optical frequency to be utilized as an additional dimension for the organization of the memory. This extra dimension makes possible direct (and random) access to the information contained in very large data bases.

We have evolved technically reasonable systems configurations for the types of applications normally performed by "direct access storage devices" (DASD). (In today's technology, DASD functions are typically performed by inductive magnetic recording on disks.) For very large centralized data bases, these configurations have favorable

price/performance compared to projected conventional technologies (Section III, Publications, No. 4, No. 5 and No. 6).

The basic concept is to utilize a centralized laser which is repetitively scanned in frequency space over the entire inhomogeneous line every 30 μsec. Writing of the information contained in one spatial storage location is accomplished by modulating the effective photochemical hole burning rate by a time varying external gating voltage while the laser frequency is scanned. This gating could be achieved, for instance, by driving a fast light gate placed in front of the recording medium or by using an external field to control the quantum yield of the hole burning process itself. For reading, the laser is FM modulated at ω_m less than or equal to the hole width and the intensity of the FM spectroscopy beat signal monitored as a function of time. Under these conditions, the hole lineshapes are effectively differentiated and the slowly varying inhomogeneous lineshape is suppressed. Pulse shaping electronics are employed to reproduce the original data pulse shapes.

The laser beam is time shared between approximately 100 identical "arms". Each arm consists of a $10^3 \times 10^3$ spot XY galvonometer driven mirror pair, a holographic optical element which acts both as a focusing lens and a 64× beam multiplexer, and an array of 64 1 cm squares of recording medium. The 100 arms are contained in a cubic meter LHe cryostat.

Each 1 cm square of recording medium contains 10^6 spatial storage locations and hence $10^3 \times 10^6$ or 10^9 bits of information. Each arm thus contains 6.4×10^{10} bits, and the entire system contains $6.4 \times .10^{12}$ bits. The data is organized into 6.4×10^4 bit pages, each defined to consist of 10^3 time domain bits flowing in the 64 parallel data channels. The time to react or write a page is thus 30 μsec.

Since galvonometer driven mirrors have settle times on the order of 3 msec, only 1% of the time for each arm is spent reading or writing data. The remaining 99% of the time is effectively dead time, while the mirrors are moving to the next position. This make possible time sharing of the centralized components among the 100 arms to insure that data is being read or written into one of the arms at all times. Thus the average data rate is on the order of 2×10^9 bits/sec. Data erasing is most easily accomplished on an arm by arm arm basis using flood illumination from a UV lamp.

D. Survey of PHB Recording Materials

One of our major materials research goals was to discover PHB recording materials sensitive to radiation in the GaAlAs laser range (λ between 750 nm and 900 nm). An actively stabilized infrared dye laser facility was set up to provide a means of rapidly testing materials with zero phonon line in this region. Using the Mollwo-Ivey relations mentioned in Part B above as a guide, the 837.3 nm line of the N_1 center in NaCl and the 833.0 nm line in R' center in LiF were investigated.

Only the R' in LiF turned out to exhibit PHB behavior (Section III, Publications, No. 7). Fortunately, this materials system is particularly convenient due to its stability at room temperature, due to the hardness and good optical quality of the host, and due to the fact that the 833 nm wavelength is at the center of the band covered by standard commercially available GaAlAs diode lasers.

Holes corresponding to relative absorption changes of as much as 10% could be burned with exposures of several J/cm^2 of infrared radiation. The most striking feature of the hole spectrum is the occurrence of the side hole to lower frequency from the burning laser and one very weak side hole to higher frequency in addition to the expected deep central hole

produced at the burning laser frequency. The central hole and side hole have linewidths on the order of 300 MHz (0.0006 nm) and the splittings between the various holes are all roughly 1 GHz. The linewidth of the inhomogeneous zero phonon line was typically 0.4 nm.

Another major topic of research was on investigation of the effects of externally applied electric fields on PHB dynamics and lineshapes (Section III, Publications, No. 8 and No. 9). No significant effect on PHB dynamics or burning speed was observed for electric fields up to 5 kV/cm. However, dramatic lineshape effects due to Stark splitting were observed. These effects were utilized to study the nature of the 607 nm color center in NaF. The previous assignment of this line to an N_1 center with C_2h symmetry was found to be inconsistent with the Stark spectroscopy results and the orientation of the permanent dipole was determined with a high level of accuracy.

E. Tunable GaAlAs Laser Sources

The wavelength tuning characteristics of several types of commercially available single transverse and longitudinal mode GaAlAs diode lasers were studied in detail. The laser beam was collimated by a microscope objective, passed through a 1.5 GHz or 30 GHz free spectral range etalon, and the transmitted intensity monitored as the laser temperature on injection current was varied.

We found that under the proper conditions, over 60 GHz of continuous, single mode tuning could be achieved simply by ramping the injection current. The bandwidth of the laser was about 200 MHz due to residual drive current fluctuations.

Since the achievable data rates are limited by the laser tuning speed, the rapid tuning capabilities of the GaAlAs lasers were extensively studied. Over 50 GHz of tuning was achieved in 25 μsec using a triangle waveform modulation of the injection current. This

corresponds to a tuning rate of 2 GHz/μsec. Assuming an idealized hole width of 50 MHz, the laser can be scanned through a hole in 25 nsec. This satisfies the 30 Mbit per second data rate requirement set forth in Part C above.

F. Reading and Writing of Photochemical Holes Using GaAlAs Diode Lasers

We have explicitly demonstrated both burning (writing) and detection (reading) of photochemical holes using current tuned GaAlAs diode lasers (Section III, Publications, No. 10). The holes were burned in the 0.4 nm wide inhomogeneously broadened 833.0 nm zero phonon line of the R' aggregate color center contained in Mg^{++} doped LiF.

Holes corresponding to relative absorption changes of as much as 5% could be burned with exposures of several J/cm^2. Typical observed hole widths were 500 MHz (0.001 nm).

Coarse wavelength tuning was provided by adjusting the temperature and fine wavelength tuning was provided by varying the injection current using specially built electronics. For writing holes, the injection current was stabilized to $\pm$0.02 mA, resulting in a measured laser jitter bandwidth of 200 MHz. For reading holes, the injection current was periodically linearly ramped in a sawtooth manner and at the same time rapidly modulated with a low amplitude, sinusoidal waveform. The ramping caused the laser frequency to repetitively scan over the spectral region containing the hole, while the sinusoidal modulation caused a rapid dithering of the laser frequency for derivative spectroscopy. (In the limit that the dithering frequency becomes greater than the laser bandwidth, the power spectrum of the modulated beam becomes similar to the FM spectrum obtained using a single frequency laser with external phase modulator.)

Fast detection of the holes was accomplished by scanning the laser over 17 GHz at a 30 kHz rate, corresponding to a tuning rate of 1 GHz/μsec. The sinusoidal modulation

frequency was 179 MHz. Relatively shallow, 500 MHz wide holes were detected in less than 500 nsec.

III. PUBLICATIONS

1. W. Lenth, C. Ortiz and G. C. Bjorklund, "Pulsed Frequency-Modulation Spectroscopy as a Means for Fast Absorption Measurements," *I. M Research Report RJ3087*, March 13, 1981 and *Opt. Lett.* **6**, 351 (1981).

   Absorption measurements are accomplished by utilizing short pulses of frequency-modulated (FM) light. The absorption is measured by detecting the heterodyne beat signal that occurs when the FM spectrum is distorted by the absorption feature of interest. By using a single short laser pulse it is demonstrated that the beat signal can build up far above the noise level within 1 μsec. The entire absorption structure can be probed by a few laser pulses. Thus pulsed FM spectroscopy permits ultrafast absorption measurements to be made by using an overall light exposure several orders of magnitude smaller than is necessary for traditional absorption techniques.

2. C. Ortiz, R. M. Macfarlane, R. M. Shelby, W. Lenth and G. C. Bjorklund, "Thin Film Aggregate Color Centers as Media for Frequency Domain Optical Storage," *IBM Research Report RJ3062*, February 20, 1981 and *Appl. Phys.* **25**, 87 (1981).

   Thin films of aggregate color centers have been produced by electron and ion irradiation of bulk alkali halide crystals. The transverse spatial distribution of the centers was controlled on a submicron scale using electron lithography. Photochemical hole burning has been accomplished for the first time in thin film of color centers, using the 6070Å zero phonon N_1 line produced by ion and electron irradiation of NaF crystals.

3. G. C. Bjorklund, W. Lenth, M. D. Levenson and C. Ortiz, "FM Spectroscopy," *IBM Research Report RJ3136*, May 13, 1981 and *SPIE* **286**, 153 (1981).

   Sensitive and rapid measurements of the absorption and dispersion associated with narrow spectral features are accomplished using frequency modulation (FM) spectroscopy. The absorption of dispersion is measured by interest. Experimental results are presented.

4. G. C. Bjorklund, W. Lenth, M. D. Levenson and C. Ortiz, "FM Spectroscopy and Frequency Domain Optical Memories," *IBM Research Report RJ3216*, August 25, 1981 and Proceedings of the Fifth International Conference on Laser Spectroscopy, Jasper, Canada, July 1981.

   There has been considerable recent interest in spectroscopic techniques involving the use of frequency (FM) laser radiation to measure the absorption or dispersion associated with narrow spectral features. In this paper, we review the basic principles of FM spectroscopy, present new results on FM excitation spectroscopy, and describe the application of these techniques to frequency domain optical memories.

5. G. C. Bjorklund and G. Castro, "Frequency Domain Optical Storage," *IBM Research Report RJ3287*, October 30, 1981. To be published as a chapter in High Density Optical Recording, A. E. Bell, ed. (Van Nostrand, 1983).

   The phenomenon of photochemical hole burning makes it possible to utilize optical frequency as an additional dimension for the organization of an optical data store. Storage densities of 10^{11} bits/cm^2 may ultimately be achieved. The basic principles of photochemical hole burning are reviewed in this paper and recent research results are presented.

6. G. C. Bjorklund, W. Lenth, and C. Ortiz, "Cryogenic Frequency Domain Optical Mass Memory," *IBM Research Report RJ3297*, November 6, 1981 and and *SPIE* **298**, 107 (1981).

   Cryogenic frequency domain optical memories based upon photochemical hole burning offer the possibility of storing data at densities of up to 10^{11} bits/cm^2. The basic principles of photochemical hole burning are reviewed. Recent results on recording materials, data reading and writing and system configurations are presented.

7. W. E. Moerner, F. M. Schellenberg and G. C. Bjorklund, "Photochemical Hole-Burning at GaAlAs Laser Wavelengths," *Appl. Phys.* **B28**, 263 (1982).

   We report the first observation of persistent photochemical hole-burning (PHB) in the 8330 A zero-phonon line of R_2' centers in LiF. This demonstrates PHB at GaAlAs laser wavelengths, and in contrast to other systems, the hole lineshape is *asymmetric*.

8. R. T. Harley and R. M. Macfarlane, "High Resolution Stark Spectroscopy of the 6070Å Colour Centre in NaF Using Spectral Holeburning," *IBM Research Report RJ3581*, August 23, 1982. To be published in *Journal of Physics C.*

   The nature of the 6070Å, colour centre in NaF has been investigated using high resolution spectral holeburning. The previous assignment of this line of an N_1 centre with C_2h symmetry was found to be inconsistent with these new experimental results. Possible alternative defect structures are considered.

9. R. M. Macfarlane, R. T. Harley and R. M. Shelby, "High Resolution Laser Spectroscopy of Colour Centers," *IBM Research Report RJ3674*, November 9, 1982 and Proceedings of the 4th Europhysical Topical Conference on Lattice Defects in Ionic Crystal, August 30-September 3, 1982, Dublin.

   The application of narrow band dye lasers to high resolution spectroscopy within the inhomogeneously broadened zero-phonon lines of colour centres is reviewed. Most work so far has centred on the use of holeburning which is exhibited by the majority of centres examined. This technique has been used to make detailed Stark and Zeeman effect measurements. In addition, optical coherent transients have been used to determine dephasing times and the results of optical phase switching and free decay experiments are discussed.

10. P. Pokrowsky, W. E. Moerner, F. Chu and G. C. Bjorklund, "Reading and Writing of Photochemical Holes Using GaAlAs Diode Lasers," *IBM Research Report RJ3784*, February 11, 1983. To be published in May 1983 *Optics Letters.*

    A current tuned GaAlAs diode laser is utilized both to burn and to detect narrow photochemical holes in the inhomogeneously broadened 833 nm zero phonon line of the R' color center in LiF. Applications for reading and writing data into frequency domain optical memories based on photochemical hole burning are discussed.

IV. TECHNICAL REPORTS

1. Report #1, "Frequency Domain Optical Storage," Gary C. Bjorklund and George Castro, December 10, 1982.

   The phenomenon of photochemical hole burning makes it possible to utilize optical frequency as an additional dimension for the organization of an optical data store. Storage densities of 10^{11} bits/cm^2 may ultimately be achieved. The basic principles of photochemical hole burning are reviewed in this paper and recent research results are presented.

2. Report #2, "Cryogenic Frequency Domain Optical Mass Memory," Gary C. Bjorklund, Wilfried Lenth and Carmen Ortiz, December 10, 1982.

   Cryogenic frequency domain optical memories based upon photochemical hole burning offer the possibility of storing data at densities of up to 10^{11} bits/cm^2. The basic principles of photochemical hole burning are reviewed. Recent results on recording materials, data reading and writing, and system configurations are presented.

3. Report #3, "High Resolution Stark Spectroscopy of the 6070Å Colour Centre in NaF Using Spectral Holeburning," by R. T. Harley and R. M. Macfarlane, December 21, 1982.

   The nature of the 6070Å, colour centre in NaF has been investigated using high resolution spectral holeburning. The previous assignment of this line to an N_1 centre with C_2h symmetry was found to be inconsistent with these new experimental results. Possible alternative defect structures are considered.

4. Report #4, "High Resolution Laser Spectroscopy of Colour Centres," by R. M. Macfarlane, R. T. Harley and R. M. Shelby, December 21, 1982.

   The application of narrow band dye lasers to high resolution spectroscopy within the inhomogeneously broadened zero-phonon lines of colour centres is reviewed. Most work so far has centred on the use of holeburning which is exhibited by the majority of centres examined. This technique has been used to make detailed Stark and Zeeman effect measurements. In addition, optical coherent transients have been used to determine dephasing times and the results of optical phase switching and free decay experiments are discussed.

5. Report #5, "Reading and Writing of Photochemical Holes Using GaAlAs Diode Lasers," by P. Pokrowsky, W. E. Moerner, F. Chu and G. C. Bjorklund, February 7, 1983.

A current tuned GaAlAs diode laser is utilized both to burn and to detect narrow photochemical holes in the inhomogeneously broadened 833 nm zero phonon line of the R' color center in LiF. Applications for reading and writing data into frequency domain optical memories based on photochemical hole burning are discussed.

V. CONFERENCE PRESENTATIONS

1. *Invited Paper.* G. C. Bjorklund, W. Lenth, M. D. Levenson and C. Ortiz, "FM Spectroscopy and Frequency Domain Optical Memories," Fifth International Conference on Laser Spectroscopy, Jasper, Alberta (July 1981).
2. *Invited Paper.* W. Lenth, G. C. Bjorklund and C. Ortiz, "Frequency Modulation Spectroscopy and Frequency Domain Optical Memories," International Conference on Lasers 1981, New Orleans, Louisiana (December 14-18, 1981).
3. *Invited Paper.* G. C. Bjorklund, W. E. Moerner and F. M. Schellenberg, "Progress in Frequency Domain Optical Memories," Conference on Lasers and Electro-Optics 1982, Phoenix, Arizona (April 14-16, 1982).
4. C. Ortiz and G. C. Bjorklund, "Laser Spectroscopy of Aggregate Color Centers in Polycrystalline Hosts," XIIth International Quantum Electronics Conference, Munich, West Germany (June 22-25, 1982).
5. *Invited Paper.* G. C. Bjorklund, W. E. Moerner, F. M. Schellenberg and P. Pokrowsky, "Recent Progress in PHB Optical Memories," Symposium on Unusual Photoactive Solids, Wasserschloss Mitwitz, West Germany (June 27-30, 1982).

VI. PERSONNEL

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